

RESEARCH PROGRESS ON THE TREATMENT OF LIVESTOCK AND POULTRY FECES CONTAINING HEAVY METAL RESIDUES BY PASSIVATOR

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Abstract. In view of the current pollution of heavy metals in livestock and poultry in China, this paper discusses bioavailability of heavy metals and the classification of passivators, as well as different types passivators on characteristics of residual heavy metal passivation in livestock and poultry waste. On this basis, the research and application of physical, chemical and biological passivators are presented. The passivating effect of different passivators on heavy metals in livestock and poultry waste is introduced, which provides a technical reference for the application of passivator to the treatment of heavy metals in livestock and poultry waste.

Keywords: *livestock wastes, passivator, heavy metal residues, bioavailability, sugarcane pulp*

Introduction

With the rapid expansion of large-scale and intensive livestock and poultry production in China, the amount of livestock manure emissions has increased rapidly. According to relevant statistics: at the end of 2010, the total excrement of livestock and poultry excrement in China was about 22.35 t (Geng et al., 2013). It is predicted that the total amount of livestock and poultry manure pollution will increase by 31% in 2020 (Wu et al., 2013). It is well known that heavy metals such as Cu, Zn, As and Cr are usually contained in livestock and poultry manure, which will cause serious pollution to the water, soil and atmospheric environment without treatment. A survey shows that 20 million hectares of cultivated land have been polluted by heavy metals, accounting for 1/6 of the total area of the country's farmland (Lu et al., 2016). The amount of grain contaminated by heavy metals amounts to 12 million tons, resulted in a direct economic loss of more than 200 billion Yuan (Cai et al., 2014). The pollution caused by heavy metals in livestock and poultry waste has posed serious threat to the ecological environment and food safety of our country, and it has become one of the main factors restricting the development of green ecological agriculture in China.

Passivator is a kind of material that has adsorption, affinity and exchange metal ions. Its main function is to convert the heavy metal ions in the organic matter into fixed and stable insoluble form, reduce the bioavailability of heavy metals in the feces of

livestock and poultry, and reduce environmental pollution. The related studies showed that the distribution coefficient of exchangeable state, carbonate binding state and iron manganese oxide binding state Pb, Cu, Cr and As in pig manure after composting was significantly decreased. Specific passivator reduced the activity of heavy metals and reduced the risk of heavy metals in livestock and poultry feces from contamination of soil and crops (Zheng et al., 2005). In this paper, the bioavailability of heavy metals, the types of heavy metal passivator in livestock and poultry manure and the new progress in the application of some passivator were introduced. On this basis, some suggestions and prospects were present to provide a technical reference for the subsequent application of passivator concerning the solutions to the problem of residual heavy metal in livestock and poultry manure.

Bioavailability of heavy metals

The researchers usually define the degree of bioavailability of heavy metals and the accumulation and toxicity of migration in the organism by referring to the bioavailability of heavy metals (*Fig. 1*). The research indicates that the bioavailability of heavy metals not only relates to the content of heavy metals, but also relates to their existing forms (Huang et al., 2010). According to the common sequential extraction method (Nemati et al., 2011; Elwell et al., 2001), the form of residual heavy metals in waste can be divided into water soluble state, organic binding state, exchangeable state, carbonate binding state, oxide bound state (iron manganese oxidation state) and residue state. Additionally, the bioavailability can be reduced in turn (Yang et al., 2010). Meng Jun used DPTA extract to explore the bioavailability of Cu, Mn and Zn in pig manure. The results showed that the effective state of Zn and Cu reached 24.05-69.84% and 24.47-69.10%, indicating that the biological availability of Cu and Zn in pig manure were higher than that in general; if not treated, pig manure would cause serious pollution to the environment (Meng et al., 2014). Dong Tongxi and other researchers explored the dynamic changes of the bioavailability of Cu, Zn, Cd and Pb in the organic manure of different livestock and poultry manure, and pointed out that the bioavailability of different heavy metals in the production of organic manure from different livestock and poultry manure was different (Dong et al., 2016). Zhang Yunqing and other researchers studied the difference of bioavailability of heavy metals in manure of different livestock and poultry and their dynamic changes over culture time by using biological pot experiment method. The results showed that the bioavailability of Cu, Zn and Cd in livestock and poultry manure was significantly lower than that of heavy metals in the calcareous and acidic soil a few months after adding livestock and poultry manure (Zhang et al., 2015). Therefore, the bioavailability of heavy metals is an important indicator to measure the seriousness of heavy metal pollution. Reducing the bioavailability of heavy metals in livestock and poultry is one of the effective measures to reduce the pollution caused by heavy metals in livestock and poultry waste. The addition of specific quantities and types of passivator can greatly reduce the bioavailability of heavy metals in livestock manure.

Classification of passivator

Usually, passivators are classified into three types according to their mechanism and material properties. These include physical passivator, chemical passivator and biological passivator (Li et al., 2018).

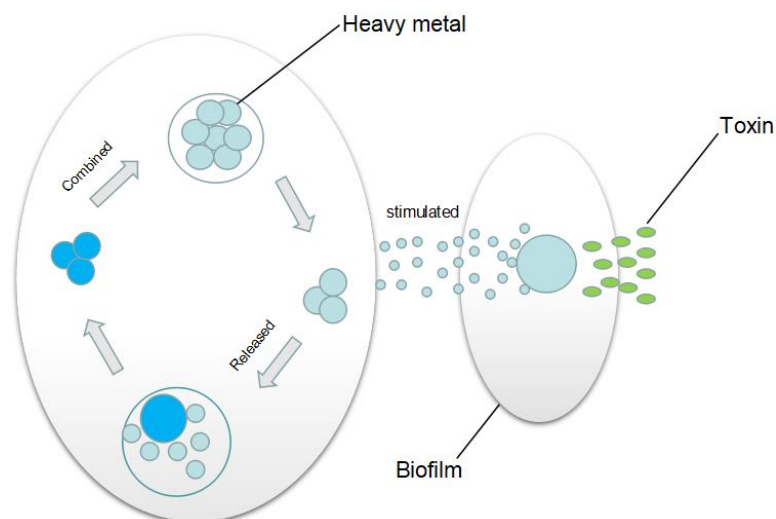


Figure 1. Schematic diagram of bioavailability

Physical passivator

In the process of organic fertilizer production, passivation means using a type of passivator to passivate heavy metals in the raw material of organic manure, as so to reduce the dissolution, transfer, bioavailability and absorption of all kinds of heavy metals (Laperche et al., 1997; Conder et al., 2001; Tang et al., 2003; Chen et al., 2006). Commonly used types of physical passivator (Fig. 2) include: dechonite, zeolite, activated carbon, sepiolite and quicklime (Li et al., 2014; Liu et al., 2017; Hou et al., 2014). Physical passivator is characterized by high porosity, large surface area, strong physical adsorption capacity and ion adsorption capacity, which can effectively reduce the biological activity of heavy metals. However, the passivation efficiency of physical passivator is not high because the passivation agent and organic fertilizer products are difficult to separate, which affects the fertilizing efficiency and quality of organic fertilizer.

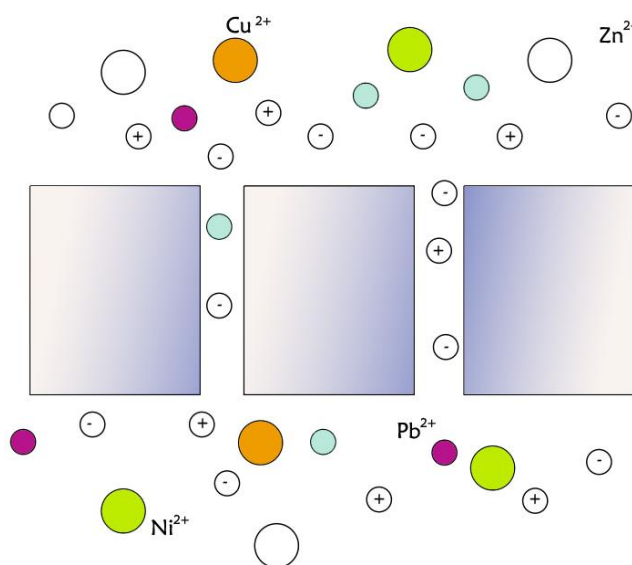


Figure 2. The adsorption mechanism of physical passivator

Chemical passivator

Heavy metal ions are mainly treated by surfactants (Zhao et al., 2003), acidifiers (Yuan et al., 2006) and organic complexing agents (Zhang et al., 2006). Usually, they can change the pH value of livestock and poultry feces by complexation, ion exchange, precipitation. Chemical passivator change the occurrence forms of heavy metals in organic fertilizer and change the bioavailability and migration of heavy metals (Liang et al., 2012). (The mechanism is as shown in Fig. 3). Chemical passivator include calcium magnesium phosphate fertilizer (Xu., 2006), phosphate rock powder (Elouear et al., 2008), fly ash (Siddiqui et al., 2004) and so on. The deficiency of chemical passivator is that it is easy to cause secondary pollution to the soil environment (Mo et al., 2001), which limits the popularization and application of chemical passivator. At present, chemical passivator are rarely used in the treatment of livestock and poultry feces, mainly because chemical passivators can damage the ecological environment. In addition, the passivator can not be recycled or decomposed into harmless substances and the recovery efficiency is low.

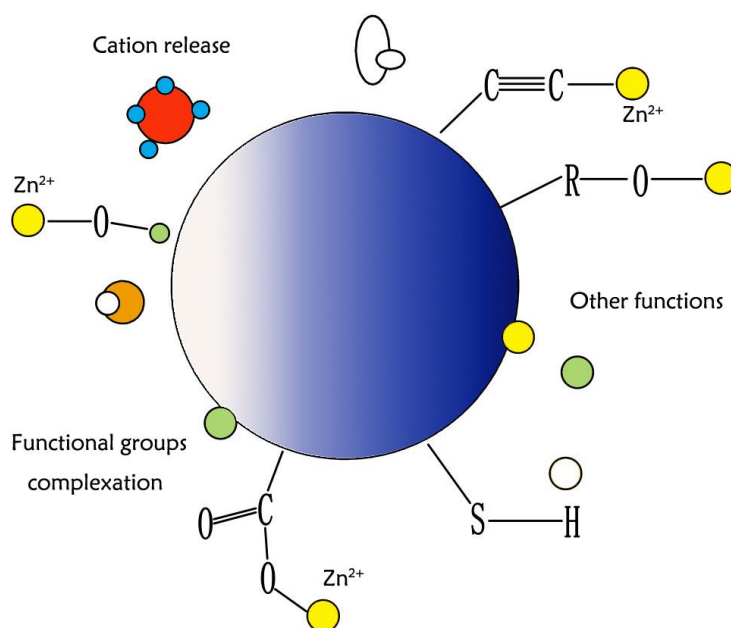


Figure 3. Schematic diagram of chemical passivation mechanism

Biological passivator

Biological passivators (Fig. 4) absorb the residual heavy metals in the waste and reduce the biological activity of heavy metals through microbial physiological action, and then form a new encapsulated biofilm and finally make it precipitate. Biological passivators mainly includes *Phanerochaete chrysosporium* (Huang et al., 2017), Chlorophyta (Anastopoulos et al., 2015), white rot fungus (Zhang et al., 2017), *Bacillus* (Wang et al., 2018) and compound biological agent (Pan et al., 2010). Studies have shown that some microbes or microbial agents such as green algae, white rot fungi and *Bacillus* can reduce heavy metals by absorption, encapsulation, precipitation, immobilization and covalent transformation of heavy metal ions in the environment (Pavel et al., 2011; Xue et al., 2012; Tian et al., 2013). In the production of organic

fertilizer, biofiltration is the main microorganism technology (He and Hu, 2011). Although the efficiency of this method is high, but the cost is too high and the post-treatment is difficult. It is easy to cause secondary pollution. At present, it is still in the stage of laboratory research and its related practical engineering application technology is not mature.

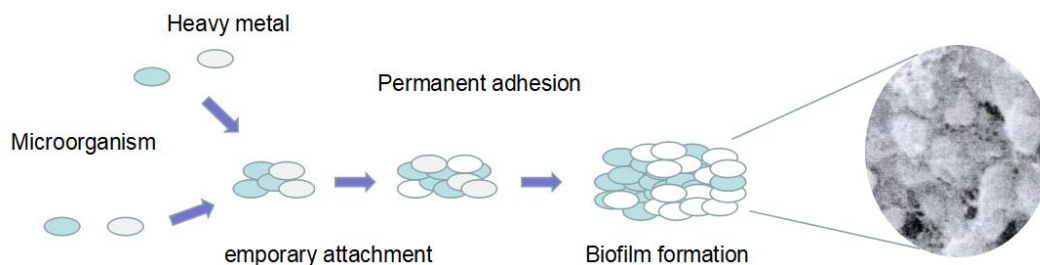


Figure 4. Microbiological affinity adsorption diagram

Any type of passivator has its own principle and characteristics. *Table 1* summarizes and compares the above passivators.

Table 1. Comparison of heavy metal passivator for livestock wastes and other wastes

Type of passivator	Principle of passivation	Common species	Advantages	Disadvantages
Physical passivator	Physical adsorption of substances with higher adsorption capacity	Activated carbon, Zeolite, Sepiolite, Bentonite, Quicklime, etc.	High porosity and large specific surface area	Passivation efficiency is low and difficult to separate
Chemical passivator	Regulating and changing the occurrence of heavy metals in compost by complexation, precipitation and ion exchange (Liang et al., 2012)	Calcium magnesium phosphate fertilizer, Phosphate rock Powder, Fly ash	Good availability and mobility	Secondary pollution of the environment limits the development of chemical passivators
Biological passivator	Fusion, adsorption, fixation, precipitation, covalent transformation	Phanerochaete Chrysosporium, Green algae, White rot fungi, Bacillus and Compound microbial inoculants	High efficiency and large handling capacity	Difficult post-treatment, biological strain contamination and currently in the experimental stage

Application of passivation agent in the treatment of residual heavy metals in animal manure

Application of physical passivator

Quicklime is a kind of traditional and effective acid soil improver, which can reduce soil pH value, reduce heavy metal activity in soil, supplement plant calcium and magnesium nutrition, increase crop yield and quality (Yu et al., 2014). Wong and Selvam pointed out that the addition of lime as passivator in sludge composting process

could effectively reduce the bioavailability of heavy metals (Wong and Selvam, 2006). Cheng Yi and other scholars analyzed the content and speciation of heavy metals in sludge before and after adding quicklime by BCR extraction method (Cheng et al., 2012). The results showed that the heavy metals in sludge could be passivated by quicklime and the contents of unstable heavy metals in sludge were decreased. Therefore, in the process of producing organic manure from livestock and poultry manure, proper addition of quicklime can effectively reduce the bioavailability of heavy metals and reduce the content of bioavailability of heavy metals in livestock and poultry manure.

Among the natural minerals found at present, Zeolite has the largest specific surface area and the best adsorption capacity (Chen, 2001). Cheng Ting and other researchers found that the specific surface area of synthetic zeolite had great influence on the adsorption of heavy metal ions Ni, Pb and Cu (Cheng et al., 2016). Li Yi and other researchers found that after the anaerobic fermentation of pig manure, the effective content of heavy metal Zn in the residue of non zeolite was 32.63%, while the effective content of Zn in the addition of zeolite was 29.12%. Therefore, zeolite addition can reduce the bioavailability of heavy metal Zn (Li et al., 2016).

Other physical passivators have also been studied for the treatment of residual heavy metals in livestock and poultry feces. Song Hefu and other researchers found that the outer surface of the small particles of sepiolite can form a hydrated oxide covering layer. Moreover, the electrical properties of the surface were negative, which is beneficial to the complexation or adsorption of heavy metal ions in the livestock and poultry manure (Song and Xia, 2000). Alvarez-Ayuso E and other scholars showed that when the amount of sepiolite was 4%, the content of available Zn in contaminated soil could be reduced by more than 95% (Alvarez-Ayuso et al., 2003).

Application of chemical passivator

Calcium magnesium phosphate fertilizer contains a large amount of phosphorus, calcium, magnesium and other elements needed for crop growth. It is a commonly used phosphate fertilizer or acid soil sample improver, which can promote the growth and development of plants and enhance their resistance to stress (Zhang et al., 2012); it can also be used as a heavy metal passivator to effectively reduce the bioavailability of residual heavy metals in compost and soil (Wu et al., 2015; He, 2011). Qian Haiyan and other researchers carried out pot experiments to study the improvement effect of calcium magnesium phosphate and lime on crop soil contaminated by Cu and Zn. The research shows that the growth of crops is basically not polluted by heavy metals in the soils with calcium magnesium phosphate and lime. Calcium magnesium phosphate can reduce the pollution of heavy metals to some extent. However, the phosphate fertilizer which is not completely absorbed can cause secondary pollution to the ecological environment (Li et al., 2017).

Phosphate mineral is a common phosphate mineral, and its main component is fluorapatite (chemical formula, $\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$). Phosphate mineral contains total phosphorus (phosphorus pentoxide) 10-35%, in which the phosphorus of 3-5% is soluble in weak acid and can be absorbed and utilized by crops, but most of the other crops are difficult to absorb and use directly. It belongs to insoluble phosphate fertilizer. Its structure has many forms. Mineral phosphorus powder is not only a passivator with high ratio of performance to price, but also does not cause environmental damage (Scheckel et al., 2003). Liu Yu and other researchers used phosphate rock to study the

adsorption of Cd^{2+} , Zn^{2+} , Pb^{2+} , Cu^{2+} , Cr^{2+} , Fe^{2+} and Hg^{2+} in livestock and poultry manure. The results showed that phosphate rock powder has a good passivation effect on most heavy metal ions (Liu and Peng, 2001).

Other kinds of chemical passivators are also used in the production of organic fertilizer. Zhang Shuqing found that in the process of pig manure composting, 10% weathered coal was added to the experiment. In the experiment, the water soluble content of Zn, Cr, Cu and As decreased 6.17%, 4.17%, and 1.83% respectively (Zhang et al., 2006). Weathered coal has passivation effect on water-soluble metal in pig manure organic fertilizer. Xi Yonghui and other researchers explored the adsorption ability of fly ash and bentonite to heavy metal ions Ni^{2+} , Pb^{2+} , Cd^{2+} and Zn^{2+} . The results showed that the adsorption capacity of Zn^{2+} is equal to that of bentonite, and that of bentonite is lower than that of fly ash. However, with the increase of heavy metal content, the adsorption effect of fly ash and bentonite on heavy metal ion Ni^{2+} , Pb^{2+} , Cd^{2+} and Zn^{2+} is decreased (Xi and Zhao, 2004).

Application of biological passivator

Phanerochaete chrysosporium is a kind of white rot fungus, which can be used to remove heavy metals (Mittar et al., 1992). Say and other showed that Phanerocephala sporophyllum had a certain adsorption ability to Cd, Pb, and Cu, but it had the strongest adsorption ability to Pb (Ridyan et al., 1999). Under the optimum adsorption conditions, the maximum adsorption capacities of Pb, Cd, Cu were 27.79 mg/g, 85.86 mg/g and 26.55 mg/g, respectively. In the process of sludge producing organic fertilizer, Wan Li Li added different proportion of compound microbial inoculants. The study showed that the compound microbial inoculum added 10‰ and the exchangeable state of Zn decreased by 17.7% (Wan, 2014). Zhou Shungui and other studies show that sludge biological leaching can effectively dissolve heavy metals in sludge by inoculating Thiobacillus ferrooxidans. After 4~10d biological leaching, the maximum removal rate of Cu, Cr and Zn can reach 80%, 100%, and 100%, respectively (Zhou et al., 2003).

Heavy metals may stimulate the growth of anaerobic bacteria during the production of organic manure. It is usually beneficial to the reproduction and growth of cells. However, high concentrations of heavy metals in animal feces inhibited bacterial growth. In particular, copper, nickel, zinc, lead and other heavy metal ions are more toxic to bacteria, even if there are only trace amounts of digestive juice, they will have serious consequences (Zhang, 1996). Xu and Guo found: in order to reduce the toxicity of heavy metals. Heavy metals can be precipitated by organic or inorganic ligands to adsorb or chelate the residual heavy metals in animal feces (Xu et al., 2018; Guo et al., 2017).

Passivation effects of different passivator on heavy metals

Different types of passivator differ in passivation effects and removal rates. *Table 2* shows the passivation effect of different passivator to heavy metal.

Conclusions and prospects

Due to the rapid development of livestock and poultry breeding industry, the environmental pollution caused by heavy metal residues in feces is becoming more and more serious. At present, the use of existing passivator can reduce the bioavailability of

heavy metal residues in livestock and poultry feces to a certain extent and can reduce the pollution of heavy metals to the ecological environment. However, there are still some heavy metals in the excrement of livestock and poultry that exceed the standard seriously, which lead to the new ecological environment pollution. The types of passivator used at present also have the disadvantages which easily cause secondary pollution and high cost. Therefore, it is necessary to further study their characteristics in order to develop a variety of green, environmentally friendly and inexpensive passivator series. In this paper, some opinions and prospects for heavy metal residues in livestock and poultry feces are presented.

Table 2. passivation effect of different passivators on heavy metals in compost

Types of passivators	Compost type	Passivator	Passivation effect (%)							Methods	References
			Cu	Zn	Cr	Pb	Cd	Ni	As		
Physical passivator	Pig manure	Meerschaum	84.9	71.2	53.8	76.2	79.3	-	89.4	Plasma emission spectrometer ICP	Liu, 2008
		Zeolite	71.0	61.6	62.9	76.3	71.0	-	79.2		
	Mud	33% shell	62.88	78.74	98.3	98.9	0.08	58.45	-	BCR extraction method	Cheng et al., 2012
Chemical passivator	Pig manure	Ground phosphate rock	48.0	51.3	53.3	63.1	51.0	-	54.0	Plasma emission spectrometer ICP	Liu, 2008
		Calcium magnesium phosphate	51.0	71.6	53.7	62.2	80.7	-	75.3		
		10% weathered coal	69.03	84.10	55.76	-	-	-	27.5	Heavy metal passivation experiment	Zhang et al., 2006
		Fly ash	-	88.40	-	86.08	89.60	66.20	-	Dynamic adsorption test	Xi et al., 2004
	Chickn manure	10% Efflorescent coal	76.91	79.34	63.93	-	-	-	24.98	Heavy metal passivation experiment	Zhang et al., 2006
Biological passivator	Mud	Thiobacillus ferrooxidans	80	100	100	-	-	-	-	Bioleaching method	Zhou et al., 2003
		Thiobacillus ferrooxidans + 4 g/L Fe ²⁺	92	83	55	16	-	54	-		Zhou et al., 2002

1) Physical, chemical and biological passivator have their own characteristics. The first two types of passivator have been used in the basic mature technology, but their comprehensive performance is not very well, it is necessary to further study the characteristics of the passivator to reduce its shortcomings. However, biological passivator has high treatment efficiency, biological strain pollution and high cost of treatment. At present, this type of passivator is still in the experimental stage. In the future, it is necessary to further improve the technology and solve the related practical

engineering problems. In particular, a series of complex biological passivators with low cost, high efficiency and no pollution have been developed by using gene technology.

2) At present, many scholars are studying the compound passivator and its performance is better than that of single passivator. In the future, big data can be used as the supporting platform and advanced mathematical methods can be used for the experimental study of the passivating agent. Compound passivator can solve the technical bottleneck of engineering so that it can be popularized and applied.

3) We should make full use of agricultural wastes rich in resources. For example, sugarcane pulp is a kind of renewable biomass material from sugarcane waste. The treatment of residual heavy metals in feces by modified sugarcane pulp can effectively improve the recovery efficiency of resources. The modified sugarcane pulp was mixed with a certain amount of quicklime and calcium magnesium phosphate fertilizer to form a compound passivator, which is more efficient than a single treatment of heavy metals in feces and greatly reduces the biological activity of heavy metals in the feces. As a compound passivator, the modified sugarcane pulp provides a technical reference for the subsequent treatment of heavy metals. The types of passivators have a great development prospect.

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